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CALCULATING ACCURATE SHUFFLER COUNT RATES WITH APPLICATIONS

Phillip M. Rinard
Los Alamos National Laboratory
Safeguards Science and Technology
Los Alamos, NM 87545 USA
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ABSTRACT

Shufflers are used to assay uranium and other fissile elements in bulk and waste quantities. They normally require physical calibration standards to achieve the most-accurate results, but such standards are generally rare and expensive, so inappropriate standards are often used out of necessity. This paper reports on a new technique that has been developed to calculate accurate count rates, in effect simulating physical standards with rapid and inexpensive calculations. The technique has been benchmarked on existing oxide and metallic standards, used to study a variety of conditions for which standards do not exist, and applied to inventory items needing verification measurements even though appropriate physical standards do not exist.

INTRODUCTION

Shufflers nondestructively assay fissile materials in a wide variety of physical and chemical forms.[1,2] Delayed neutrons are counted after fissions are induced by a ^{252}Cf neutron source that "shuffles" between a shield and the assay chamber. The excellent precision of a count rate is a strength of shufflers, while accuracy depends on the quality of the calibration. Shufflers can be used on any fissile material, but they are most commonly used on uranium materials because plutonium is usually better assayed with a passive neutron technique; an exception is when other passive emitters (such as ^{244}Cm) are mixed with the plutonium.

There are few certified calibration standards available, generally U_3O_8 and PuO_2. But for a decade, a popular shuffler design for 55-gallon drums (Fig. 1) has been used to assay many other compounds and isotopes, such as metals, carbides, oxide mixed with graphite, ^{233}U, and ^{237}Np. Using the U_3O_8 calibration on these (or any other) materials induces a large bias to the resulting uranium mass. It is not realistic to think that new standards will be created for most (if any) of these other materials because of the cost involved, so a method for calculating accurate shuffler count rates has been developed to assist in calibration or bias correction.

The shufflers used to collect the data referred to in this paper are three instruments of the same Los Alamos design, fabricated by Canberra Industries, and installed at Los Alamos and Lawrence Livermore national laboratories. Figure 1 is a photograph of such a shuffler prior to installation.

Fig. 1. This is a shuffler designed to assay items as large as 55-gallon drums. The assay chamber is behind the doors on the lower half of the shuffler. The upper half is a storage shield for the ^{252}Cf source when it is not irradiating an item.

CALCULATIONAL TECHNIQUE

The basic mathematical description of shufflers is developed in Ref. 1, where Eq. (17) gives the number of delayed neutrons *D* produced during an assay when the irradiating flux and fission rate are constant. For the shuffler shown in Fig. 1, the flux is usually not constant because the ^{252}Cf source scans vertically during each irradiation. This is done to produce a uniform irradiation throughout the assay chamber, which is large enough for a 55-gallon drum. However, that equation can be slightly rewritten for any flux, constant or not.

$$D = M_{DN} \cdot e \cdot \sum_{j=1}^{6} P_j(t_i) \cdot \left(e^{-l_j \cdot t_r}\right) \cdot \left(1 - e^{-l_j \cdot t_c}\right) \cdot \left[\frac{n - (n+1) \cdot e^{-l_j \cdot t} + e^{-(n+1) \cdot l_j \cdot t}}{\left(1 - e^{-l_j \cdot t}\right)^2}\right]. \qquad (1)$$

The parameters have these meanings:

M_{DN} = neutron multiplication involving fissions induced by delayed neutrons,

ε = detection efficiency for delayed neutrons,

j = index of the six groups of delayed neutron precursor nuclei,

$P_j(t_i)$ = number of delayed neutron precursors for group j after an irradiation time t_i, when the number is zero at the start of the irradiation,

λ_j = decay constant for precursor group j,

t_r = time used to return the ^{252}Cf source to the shield after an irradiation,

t_c = delayed neutron count time after a single irradiation,

τ = cycle time between successive irradiations, and

n = number of irradiation and count cycles used in the assay.

The delayed neutron multiplication and detection efficiency must be calculated for the particular item being assayed, and Monte Carlo techniques are used for this purpose. The other multiplication involving fissions induced by ^{252}Cf neutrons is included in the other Monte Carlo calculations for $P_j(t_i)$ using the code MCNP.

The evaluation of $P_j(t_i)$ is done by numerically solving the basic equation for the growth of the population of delayed neutron precursors during an irradiation.[1]

$$\frac{dP_j}{dt} = f(t)\cdot\beta_j - \lambda_j \cdot P_j(t),\ j = 1, 2,...,6. \quad (2)$$

The β_j are the fractions of the fission neutrons that are delayed in the six precursor groups. The fission neutron production rate *f(t)* is a function of time, unlike the simpler case in Ref. 1, because the source is moving. The rate *f(t)* is closely related to the probability $p_{fission}(t)$ of a neutron from ^{252}Cf inducing a fission in the fissile material.

$$f(t) = p_{fission}(t)\cdot Y \cdot \bar{u} \ . \quad (3)$$

Y is the neutron yield (neutrons per second) from the ^{252}Cf source and $\bar{n}$ is the average number of neutrons released per fission. Some fission probabilities $p_{fission}(t)$ at selected selected times (and corresponding positions) of the ^{252}Cf source are calculated with the Monte Carlo code MCNP. To get $p_{fission}(t)$ at any time requires the interpolation among these calculated probabilities.

The scanning protocol specifies where the ^{252}Cf source is as a function of time during a single irradiation. In effect, a table is generated that gives $p_{fission}(t)$ at a set of closely spaced times. We also know *f(t)* numerically from Eq. (3). Equation (2), which is really six equations, can now be solved numerically starting with $P_j(0) = 0$ and working up to the end of the irradiation at t_i. The simplest numerical integration technique has worked very well.

$$P_j(t+\Delta t) \approx P_j(t) + \Delta P_j(t) = P_j(t) + \left(f(t)\cdot b_j - l_j \cdot P_j(t)\right)\cdot \Delta t \ . \quad (4)$$

The fourth-order Runge-Kutta algorithm was also implemented and tested, but the results were identical to those from Eq. (4), so the simpler procedure continues to be used. A practical time interval is 0.001 s.

All of these calculations, except for the MCNP runs, are done with the VisualBasic tools built into Microsoft's EXCEL spreadsheet. The nuclear and shuffler parameters are entered into a spreadsheet and VisualBasic "macros" generate MCNP input files, read MCNP output files, and solve Eq. (4) and then Eq. (1).

If only one fissile isotope dominates in one or more MCNP cells, a fission tally can be used to get $p_{fission}(t)$ and the delayed neutron parameters for that isotope can be used in the equations. But if more than one significant fissile isotope is present in an MCNP cell, the tally gives the fission probability for the combination of fissile isotopes and it is not clear what nuclear parameters should be used. In that case, the tally is not used. MCNP output files provide fission information on individual isotopes summed over all cells, and this is used to get $p_{fission}(t)$ for each fissile isotope. A count rate for each isotope is calculated using Eqs. (4) and then (1); these are summed to give the measured rate. A side benefit of this approach is that we can see how much each isotope contributes to the count rate.

The multiplication and detection efficiency in Eq. (1) vary slightly with the material being measured, so for the most accurate results, they should be calculated for each item, again using MCNP. The detection efficiency needs only a single, short calculation, but the multiplication takes two short calculations with and without fission neutron tracking of tallies of fission rates and products of fission rates with the number of fission neutrons produced. One of the multiplication calculations can be done at the same time as a $p_{fission}$ calculation, so only one extra calculation is needed for the multiplication.

ACCURACY TESTS

The calculational technique outlined in the previous section was tested in several ways and found to be accurate. The first test used a stationary ^{252}Cf source to study the simple case of constant flux. A can with U_3O_8 was irradiated and released delayed neutrons. The source was quickly moved to a fixed position, left in place for a number of seconds, and then quickly returned to the shield so that delayed neutrons could be counted. A total of 25 different positions, one inch apart, were used to generate a profile of $p_{fission}$ versus ^{252}Cf position. The calculated and measured values were the same within the experimental uncertainties.

The scanning protocol with its time-dependent flux was then used for the rest of the tests, starting with eight certified U_3O_8 standards, two uncertified higher-mass U_3O_8 items, U and Pu metal in weapon pits (nine pits of five types were measured),[3] and stacks of highly enriched uranium (HEU) metal disks that formed 6-cm-diameter cylinders with ^{235}U masses from 244 to 3684 g. All of these materials are very well characterized. The major uncertainty in the calculational process is the neutron yield of the ^{252}Cf source. It is known approximately (±10%) from the fabricator's documentation, but we are striving for much better accuracy. It is also crucial that the same yield accurately calculates count rates for all the different materials listed above; if this were not true, there would be a major flaw in the calculational technique. A secondary uncertainty is the density of the U_3O_8 powder; a higher density reduces the count rate. We can say that for these three types of standards there are two free parameters: the neutron yield and the density of the oxides. (Measurements on these oxides were done with a uniform handling technique to avoid changes in density.)

A single ^{252}Cf mass was indeed found that gave accurate count rates for all these materials. The HEU metal disks with well-characterized materials and a simple geometry gave a ^{252}Cf mass of 443 ± 3 μg. The pits gave a mass of 442 ± 10 μg. The oxides gave a mass of 443 ± 7 μg when the density was 4 g/cm^3. (The actual oxide density is unknown, but 4 g/cm^3 is quite plausible.) The ^{252}Cf mass is taken to be 443 μg. Of course, this mass applies only to the two shufflers at Los Alamos on decay reference dates that were chosen long ago to make the two ^{252}Cf sources produce the same count rates; other shufflers have different ^{252}Cf masses and decay reference dates.

Figures 2 and 3 compare the measured and calculated count rates for the oxides and metal disks. The oxides that gave the two points with the highest masses in Fig. 2 are not certified standards, but apparently their declared masses are fairly accurate. All the other data on the oxide curve involve certified standards. Figure 4 compares count rates for the pits and shows excellent agreement between calculated and measured values, even when more than one fissile element was present.

The calculational method is seen to be very accurate for all three types of materials. In the process, the mass of the ^{252}Cf source was determined, as was the density of the oxides.

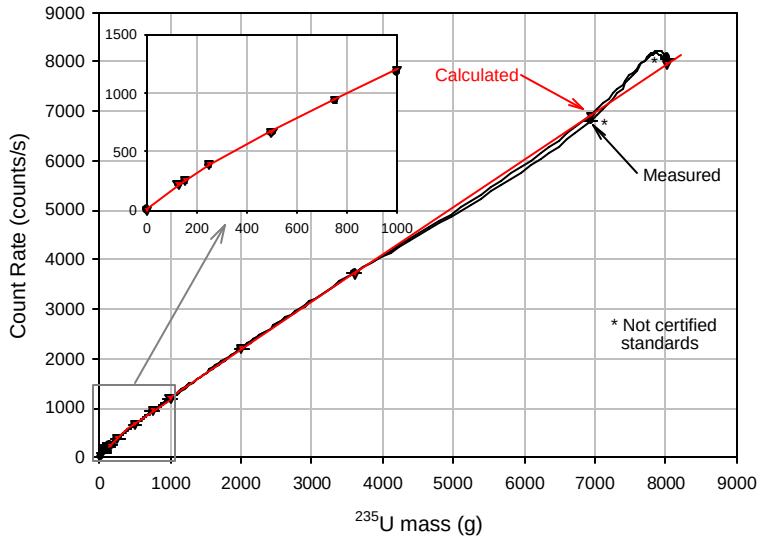

Fig. 2. Measured and calculated count rates are compared for a wide range of ^{235}U masses in U_3O_8. The inset is an enlargement of the low-mass region. The data points of measured and calculated rates at and below 3600 g-^{235}U too nearly coincide to be easily distinguished.

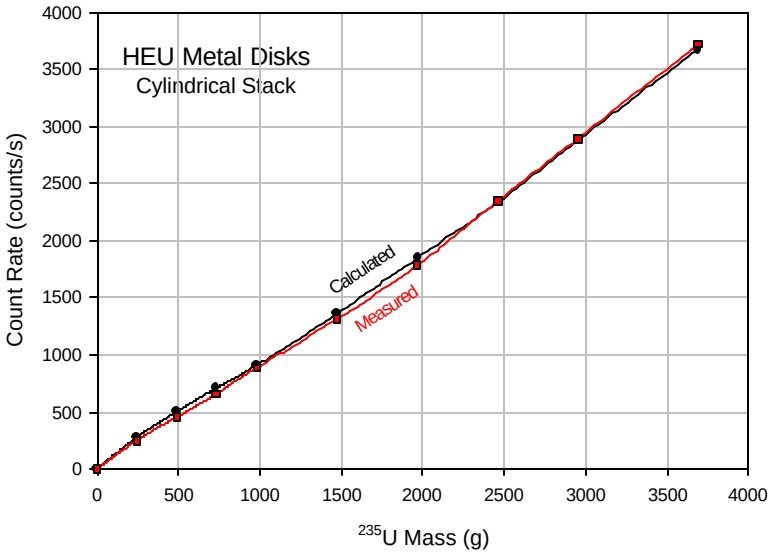

Fig. 3. Measured and calculated count rates are compared for cylindrical stacks of HEU metal disks.

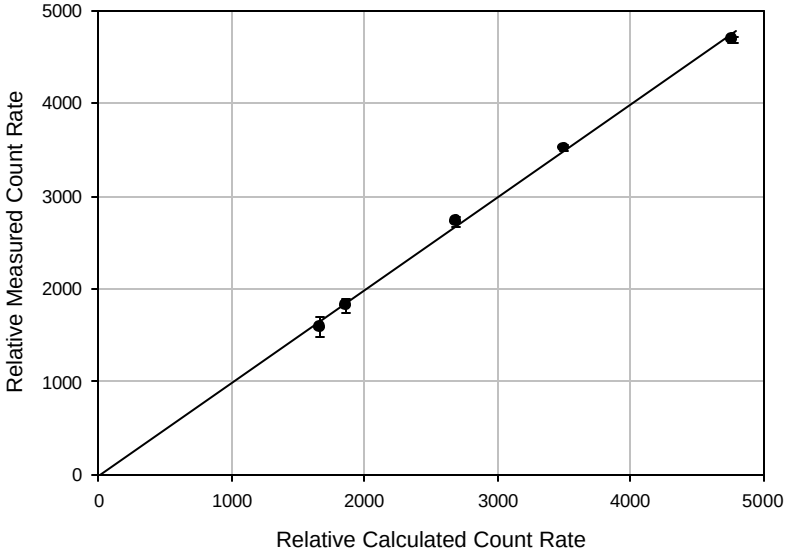

Fig. 4. Measured and calculated count rates (in relative units) are compared for nine pits of five types.

APPLICATIONS

With the calculational technique proven to give accurate count rates for standards, it can be used to explore materials and conditions that are difficult, expensive, or even impossible to create and measure directly. Making physical standards to generate the data presented below would cost many millions of dollars, assuming a fabricator could and would even take on the work. Yet these materials are commonly found in Department of Energy inventories.

Oxide Density

It was mentioned earlier that the density of an oxide affects the count rate. This can now be quantified by calculating count rates with different densities. Figure 5 shows the case of 1000 g-^{235}U in U_3O_8 (94% enriched) in a can with a radius of 6.3545 cm. There is about a 10% drop in count rate over the large density range from 2 to 10 g/cm^3.

This shows us that if the density is known only as 4 ± 1 g/cm^3, the uncertainty introduced into the calculated count rate is less than 2%. Unless the density is extremely low, an error in the assumed density has little consequence because the curve is fairly flat beyond 5 g/cm^3. Measurements of actual cans of oxide show the same small, but real, density effect. The density can be changed by shaking the powder or tapping the can on a table top.

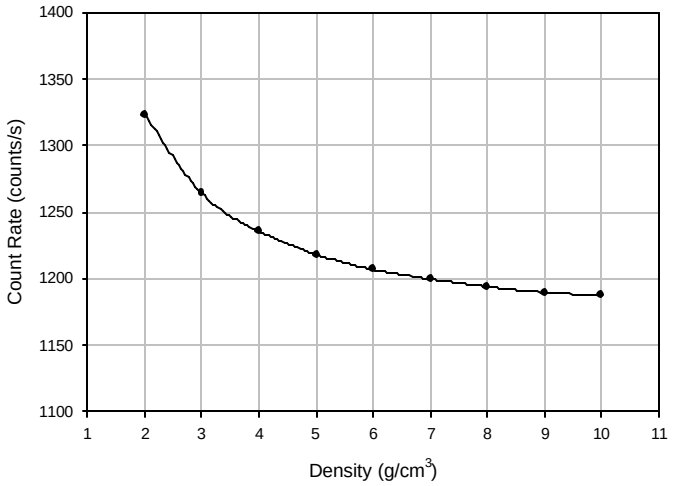

Fig. 5. Calculated count rates for 1000 g-^{235}U in U_3O_8 (94% enriched) are shown for densities from 2 to 10 g/cm^3. The radius of the material stays constant, so the fill height varies.

Metal Density

The density of metals also can vary, although not as much as with oxides. Phase changes and microcracking are two sources of density changes. To examine density's impact on count rate a sphere with 1000 g-^{235}U (94% enriched) was given different radii and densities. Figure 6 shows the resulting count rates.

Changing from 19 g/cm^3 to 16 g/cm^3 has only about a 1% impact on the count rate. This small change is readily apparent in Fig. 6 with its expanded count rate scale, but when all sources of uncertainty in a calculated count rate are considered, a 1% effect is not significant. The density of a metal item is not likely to be important.

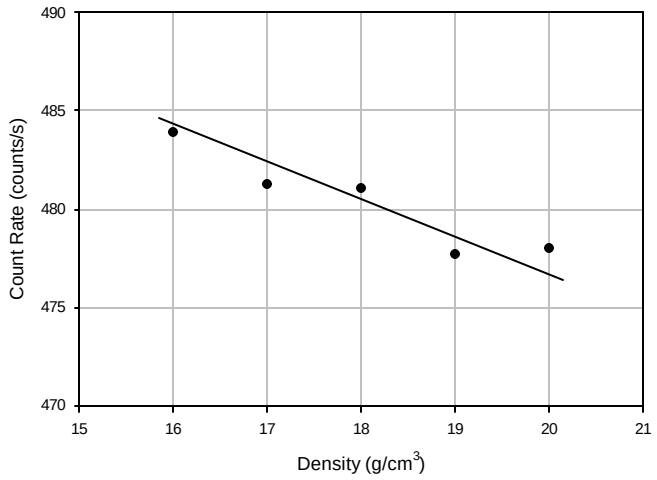

Fig. 6. Count rates are shown for spheres with 1000 g-^{235}U (94% enriched), but different radii and densities. The change in count rate is quite small and the spread about the line shows the small uncertainties in the calculations.

Oxide Moisture

The moisture content in oxides can vary according to the details of processing. The hydrogen in water is an important neutron moderator and will affect fission and count rates. Figure 7 shows the change in count rate for a can of U_3O_8 with 500 g-^{235}U (94% enriched) as the moisture fraction is varied. The volume of the oxide was kept constant on the assumption that the moisture fills voids among the oxide grains. The density of the dry oxide was taken to be 2 g/cm^3.

In this example there is a 7% increase in the count rate when the moisture content changes from 0% to 4%. Multiplication appears to grow rapidly at 10%. Moisture effects can be much more important than density effects and can cause errors in calculated rates well outside measurement uncertainties.

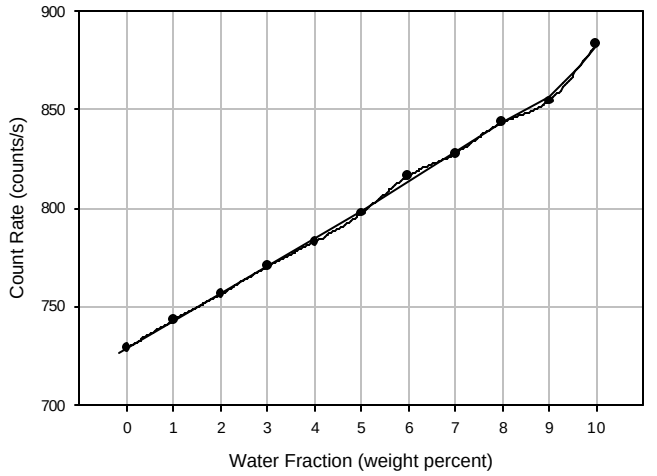

Fig. 7. The importance of moisture in oxides was studied with 500 g-^{235}U in U_3O_8 (94% enriched).

^{235}U Enrichment

Enrichment is important because ^{238}U fissions (by neutrons only with energies above 1 MeV) and absorbs neutrons without fissioning. The effect of enrichment was studied with 1000 g-^{235}U in U_3O_8 with a density of 4 g/cm^3. The enrichment was changed by adjusting the amount of ^{238}U present. The results are shown in Fig. 8.

At 100% enrichment there is no ^{238}U, so the count rate of 1214 counts/s is from ^{235}U alone. As ^{238}U is added, the enrichment decreases. With an enrichment of 10%, there is 9 kg of ^{238}U in addition to the 1 kg of ^{235}U and the count rate is 75% greater than with 100% enrichment because of all this ^{238}U. But in the HEU region between 90% and 100% enrichment, the variation in count rate is less than 1%. We learn from this that errors in the enrichment within the HEU range are unimportant unless the calibration is accurate to less than 1%.

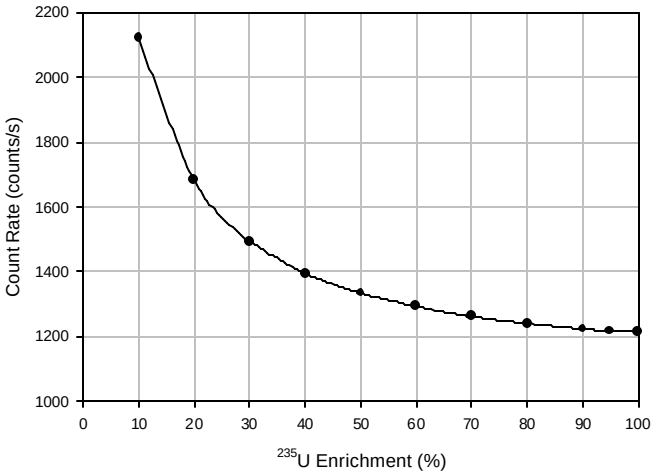

Fig. 8. A can with 1000 g-^{235}U in U_3O_8 with density 4 g/cm^3 was given different enrichments by changing the amount of ^{238}U.

Oxide Shape

The same oxide could be put in cans of different sizes. A larger-diameter container will have a smaller fill height for the same mass of oxide. This is likely to reduce the self-shielding by spreading the material into a thinner shape and increase the probability that delayed neutrons will escape the oxide. A larger count rate is expected for a larger-diameter can.

This was tested with calculations on 500 g-^{235}U in U_3O_8 (94% enriched) with a density 2 g/cm^3. The oxide was mathematically put into cans with diameters of 5 and 7 in., giving fill heights of nearly 1 and 2 cm, respectively. The larger diameter can gave a count rate higher by only 1.2%. A rather large 40% increase in diameter resulted in a nearly negligible change in count rate. The fill heights differed by a factor of two, but they were both still thin compared to the diameter. A much smaller diameter can would show a bigger effect, but the two sizes chosen here are more typical of those in use.

Metal Shape

Metallic uranium has a wide variety of shapes and a particular shape is not always known unless the can is opened or radiographed. How large are changes in count rates caused by different shapes? This was studied using 2000 g-^{235}U of metal (94% enriched) with a density of 19 g/cm^3. The results are shown in Fig. 9.

A sphere has the smallest surface area and therefore the most self-shielding; its count rate is the lowest of all shapes. A cylinder with the height equal to the diameter or even twice the diameter gives only a slightly greater count rate than the sphere. A cube or a rectangular prism has a slightly higher count rate yet. Making nine smaller spheres out of the material and packing them closely together has still less self-shielding, but the count rate is only 5% larger than the single sphere's. So far this is good news because the shape has little impact.

But the large, hollow, spherical shell has almost double the count rate of the solid sphere because self-shielding is almost nonexistent and simply because the larger size increases the ^{252}Cf neutron flux through it.

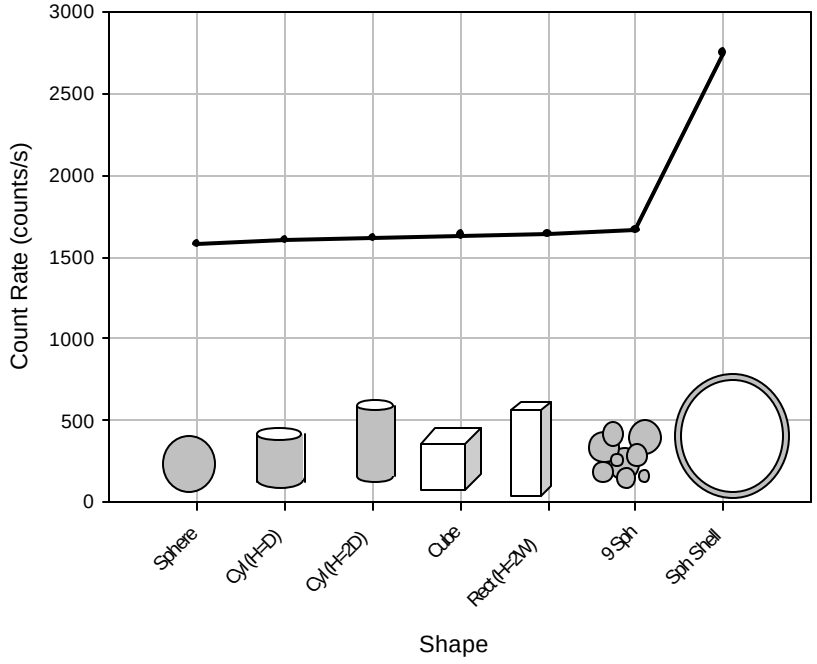

Fig. 9. The same metallic 2000 g-^{235}U (94% enrichment) was given the shapes shown here. The variations in count rate were 5% or less as long as the metal had a solid shape or consisted of closely packed small spheres. Only when a large, mostly empty shape was used did the count rate greatly change.

Some cans known to contain uranium metal were measured with a Los Alamos shuffler. The calibration for U_3O_8 oxides gave ^{235}U masses lower than declared for the metals, as expected, because of the greater self-shielding. But we wanted to demonstrate quantitatively through calculations that the measured count rates were consistent with the declared uranium metal. To get information on the shapes of the metal pieces, we radiographed them. Some were irregular chunks piled in a can and were treated as a cylinder in the calculations. Thanks to the results shown in Fig. 9, we knew that the error from using this simplified shape would be small. Another object was a cylindrical annulus, and approximate dimensions could be taken from the radiograph and used in the calculations. The result was that the biases introduced by the oxide calibration for these metal pieces with an oxide calibration were quantitatively explained and the declared masses were verified.

Chemical Compounds

Fissile materials are commonly bound with other elements to form compounds. Although these other elements do not fission, they affect neutron transport and moderation. Hence, they affect fission and count rates. To see how large these effects on count rate can be, 500 g of either uranium (94% enriched) or plutonium (6% ^{240}Pu) were used in the compounds shown in Fig. 10.

Count rates for uranium in compounds differ only slightly from those of pure uranium, with the chlorine compounds having the largest impact of 3%. There is more variation among the plutonium compounds, but most of the changes are less than 4% with the largest change being 7%. While the effects of compounds are small, they are easily measured and should not be ignored.

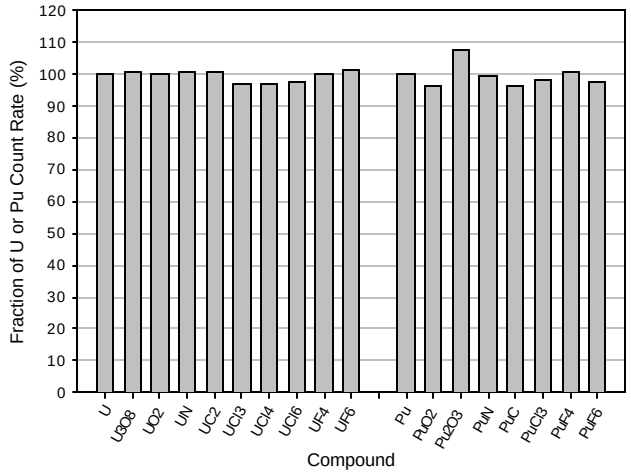

Fig. 10. Common chemical compounds of uranium and plutonium were used to determine the importance of nonfissile elements on the count rate. This figure shows the relative changes in count rate from pure uranium or plutonium.

CONCLUSIONS

The ability to calculate accurate shuffler count rates has resulted in many new opportunities for studying shuffler applications and improving shuffler meaurements. Materials can be simulated by the calculational technique for which physical standards do not or never will exist. For example, it is easy to change density or moisture content for the calculations, but this would be difficult and expensive to accomplish with physical standards.

The calculational technique has already been applied to a few actual cases where items in uranium inventories had to be assayed without appropriate calibration standards. Some were metals, some were oxides mixed with graphite, and some were oxides different from the calibration standards. The calculations could be considered as calibration points or more cautiously used to calculate bias corrections so that an established calibration can be used on other materials.

In the past, many measurements would have profited from this tool and we anticipate applying it to many inventory measurements in the future.

ACKNOWLEDGEMENTS

The data used to develop and test the calculational technique were taken over several years by Jon Hurd and Faye Hsue of Los Alamos National Laboratory and Mark Mount (and coworkers) at Lawrence Livermore National Laboratory. Their cooperation and feedback made the work reported here possible.

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